INELASTIC NEUTRON SCATTERING STUDY OF CARBON NANO-MATERIALS

A.I. Kolesnikov and C.-K. Loong

Argonne National Laboratory, Argonne, IL 60439, USA

A.P. Moravsky and R.O. Loutfy

MER Corporation, Tucson, AZ 85706, USA

Keywords: Neutron scattering, carbon nanotubes, fullerenes.

Abstract

Vibrational spectra for single wall carbon nanotubes, double wall carbon nanotubes, single wall carbon nanohorns and C_{60}-peapods have been measured with inelastic neutron scattering in a wide range of energy transfer, 5-220 meV. A decrease in intensity around 75-100 meV and the appearance of two peaks around 120-125 meV and 150 meV in the double wall nanotubes and C_{60}-peapods spectra compared to single wall carbon nanotubes and nanohorns were observed. These findings indicate the possibility of strong interaction between the walls of the double wall carbon nanotube, and between C_{60} molecules and carbon nanotube of the peapod.

RESULTS AND DISCUSSION

Although the existence of varieties of carbon nano-materials has been known for some time, the investigation of their vibrational spectra was mainly limited to the use of optical spectroscopy. In spite of the well-recognized advantages of inelastic neutron scattering (INS) technique, namely, unrestricted by the selection rules of infrared and Raman spectroscopy and accessible to all vibrational modes in the reciprocal space, so far this method was used for the study of only fullerens (see *e.g.* review in [1]) and single-walled carbon nanotubes (SWNT) [2,3]. Here we present the INS measurements on a series of carbon nano-materials prepared by MER: SWNT (~14 Å diameter), double-walled carbon nanotubes (DWNT of ~25 Å diameter), carbon nano-horns and fullerene-C_{60} encapsulated into SWNT (so called C_{60}-peapods), aiming at the characterization of the vibrational spectra over a wide energy scale, from collective motions to localized atomic vibrations.

The spectrum of nano-horns is similar to that of SWNT (see figure 1), and the latter one agrees with the main features reported in the literature. The INS spectrum of C_{60}-peapods and DWNT are very different compared to the spectra of both pure SWNT and pristine or polymerized fullerenes. The spectral intensities of the DWNT and C_{60}-peapods show a strong reduction in the "breathing" modes (normal to surfaces of C_{60} or SWNT "molecules") over the range of 30-100 meV, and an increase at higher energies toward the "tangential" modes around 110-160 meV; exhibiting new excitations at about 120-125 and 150 meV.

A possible explanation of the observed new peaks at ~120-125 meV and ~150 meV in the spectra of generalized vibrational density of states, *G(E)*, for DWNT and C_{60}-peapods is a strong change in the C-C interaction between the inner and outer walls in DWNT and between the C_{60} and nanotube in C_{60}-peapods. Some theoretical models (see *e.g.* [4]) predict the changes of two-dimensional sp^2 bonds in cylindrical "graphene" layers in nanotubes of large diameter to three-dimensional sp^3-like bonds in nanotubes of smaller diameter. A possible change of sp^2 to $sp^{2+\delta}$ ($0<\delta<1$) hybridization in DWNT and C_{60}-peapods would result in spectral changes observed in the present work. The features at energies 120-175 meV in the *G(E)* spectrum for diamond [5] (with only sp^3 bonds) show similarities with the spectra for DWNT and C_{60}-peapods (see figure 1).

Work performed at Argonne National Laboratory is supported by the U. S. DOE-BES under contract No. W-31-109-ENG-38.

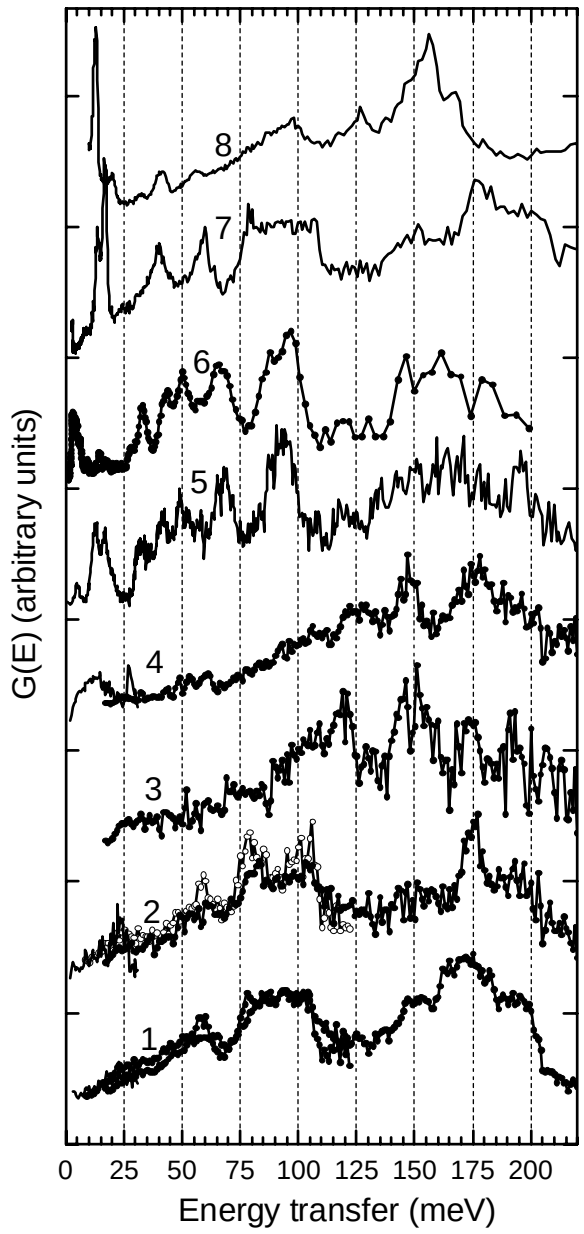

Figure* 1. *Generalized vibrational density of states, G(E), obtained from INS spectra for different carbon nano-materials: 1 – SWNH, 2 – SWNT, 3 – DWNT, 4 – C_{60}-peapods, 5 - high-pressure polymerized C_{60}, 6 - pristine C_{60}, 7 – graphite, and 8 – diamond. The spectra 1 to 4 were measured at T=8 K on the direct-geometry HRMECS spectrometer (IPNS, ANL) with different incident neutron energies, E_i=280 meV (1-4); 140 meV (1 and 2) and 50 meV (1, 2 and 4). The spectra for pristine and polymerized C_{60} [6], and graphite and diamond ([5], the data have been taken from TFXA database) are shown for comparison.

REFERENCES

1. Pintschovius, L., "Neutron Studies of Vibrations in Fullerenes," *Rep. Prog. Phys*. **57**, 473 (1996).
2. Rols, S., et al., "Phonon Density of States of Single-Wall Carbon Nanotubes," *Phys. Rev. Lett*. **85**, 5222 (2000).
3. Sauvajol, J.L., et al., "Phonons in Single Wall Carbon Nanotube Bundles," *Carbon* **40**, 1697 (2002).
4. Ding, J.W., Yan, X.H. and Cao, J.X., "Analytical Relation of Band Gaps to Both Chirality and Diameter of Single-Wall Carbon Nanotubes," *Phys. Rev*. **B66**, 073401 (2002).
5. Walters, J.K., et al., "A Spectroscopic Study of the Structure of Amorphous Hydrogenated Carbon," *J. Phys.: Condens. Matter* **7**, 10059 (1995).
6. Kolesnikov, A.I., et al., "Neutron Scattering Study of a High-Pressure Polymeric C-60 Phase," *J. Phys.: Condens. Matter* **8**, 10939 (1996).